2015 年度 修士論文要旨

Inter-molecular Interaction of Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) Studied by Low-Frequency Raman and Terahertz Spectroscopies

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Poly(3-hydroxybutyrate) (PHB) is the most studied linier polyester from PHAs group which accumulated in various microorganisms. Its ability to be degraded naturally shows a valuable potential in various applications as alternative to conventional fuel-based polymers. However, the highly crystalline of PHB make it stiff and brittle to be applied. In order to overcome this problem, it has been copolymerized with poly(3-hydroxyvalerate) (PHV), giving rise to P(HB-*co*-HV). The higher order structure of P(HB-*co*-HV) is determined by two types of inter-molecular hydrogen bonding interactions (Fig). In spectroscopic study, information related to this hydrogen bonding can be observed in the low-frequency wavenumber region (330 – 3.3 cm^{-1}). In this study, we have investigated the inter-molecular interaction of P(HB-*co*-HV) in the low-frequency vibrational mode by using Raman and Terahertz spectroscopies. Two different types of samples, isotactic and highly stretched film samples were prepared for composition, temperature, and polarized dependence measurements. Inter-molecular hydrogen bonding interactions were investigated both in the high-frequency region and low-frequency region. For PHB and its copolymers, the bands appear in the 50-150 cm^{-1} vibrational region are correspond to the inter-molecular vibrational mode of C-H···O=C hydrogen bonding and intra-molecular vibrational mode of spring like motion in the helical structure. The spectra changed dramatically as increasing HV contents due to the lattice structure change. During heating process, the shift of several peaks was observed due to the weakening of inter-molecular hydrogen bonding. The polarized THz spectra of highly oriented film sample described the existence of inter-molecular hydrogen bonding more clearly. Finally, by combining polarized and temperature dependence results, the peak assignment could be estimated.

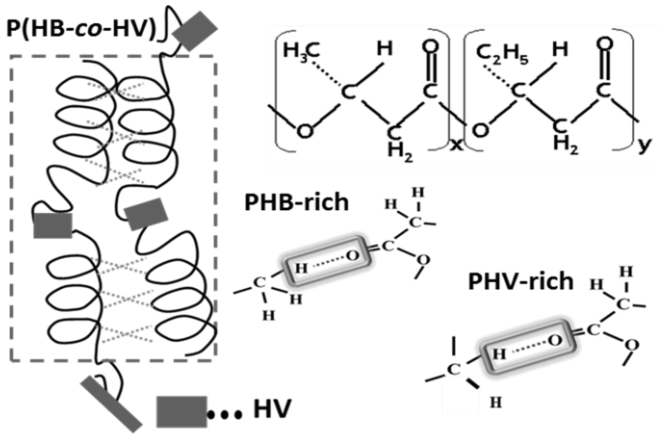

Fig. Inter-molecular hydrogen bonding of P(HB-*co*-HV)